



# **TECHNICAL REPORT ARLCD-TR-84006**

# DYNAMIC MECHANICAL PROPERTIES OF CANDIDATE LOVA AND NITROCELLULOSE BASE GUN PROPELLANTS AFTER UP TO 18 MONTHS OF ACCELERATED (HIGH TEMPERATURE) AGING

J. PINTO

D. GEORGEVICH

S. NICOLAIDES

D. A. WIEGAND

**MAY 1984** 



# U.S. ARMY ARMAMENT RESEARCH AND DEVELOPMENT CENTER

LARGE CALIBER WEAPON SYSTEMS LABORATORY

DOVER, NEW JERSEY

APPROVED FOR PUBLIC RELEASE; DISTRIBUTION UNLIMITED.

The views, opinions, and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy, or decision, unless so designated by other documentation.

Destroy this report when no longer needed. Do not return to the originator.

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE	READ INSTRUCTIONS BEFORE COMPLETING FORM
REPORT NUMBER Technical Report ARLCD-TR-84006  2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
DYNAMIC MECHANICAL PROPERTIES OF CANDIDATE LOVA AND NITROCELLULOSE BASE GUN PROPELLANTS AFTER UP TO 18 MONTHS OF ACCELERATED (HIGH TEMPERATURE) AGING	5. TYPE OF REPORT & PERIOD COVERED Final April 1981-December 1982 6. PERFORMING ORG. REPORT NUMBER
J. Pinto, D. Georgevich, S. Nicolaides,	8. CONTRACT OR GRANT NUMBER(s)
and D.A. Wiegand	
PERFORMING ORGANIZATION NAME AND ADDRESS  ARDC, LCWSL	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
Energetic Materials Div [DRSMC-LCE-D(D)] Dover, NJ 07801	AMCMS 612603.H180011
1. CONTROLLING OFFICE NAME AND ADDRESS	12. REPORT DATE May 1984
ARDC, TSD STINFO Div [DRSMC-TSS(D)] Dover, NJ 07801	13. NUMBER OF PAGES 27
14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)	15. SECURITY CLASS. (of this report) Unclassified
	15a. DECLASSIFICATION/DOWNGRADING

Approved for public release; distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

LOVA propellants

Low vulnerability propellants

Insensitive propellants

Nitrate ester propellants

Nitramine RDX propellants

Cellulosic binders

Thermoplastic elastomer binders

Surveillance

(cont)

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Several candidate low vulnerability (LOVA) propellants and several nitrocellulose (NC) base (single, double, and triple) propellants have been subjected to accelerated aging at  $65.5^{\circ}$ C and mechanical properties measured. The propellants were investigated in compression after 0, 1, 2, 3, 6, 12, and 18 months. Measurements were made at  $-45^{\circ}$ C and  $23^{\circ}$ C at a strain rate of approxi-

(cont)

# 19. KEY WORDS (cont)

Accelerated aging
RDX propellants
Compressive stress
Gun propellants
Mechanical properties
Compression
Fast strain rate

Dynamic compression
Low temperature
Ductile-brittle transition
Single-base propellant
Double-base propellant
Triple-base propellant

## 20. ABSTRACT (cont)

mately  $10/\mathrm{sec}$ . Little or no change in the compressive strengths of the candidate LOVA propellants was observed. In contrast, significant decreases in the compressive strengths of double and triple base NC propellants were found. The candidate LOVA propellants thus are more thermally stable than the NC base propellants for the conditions of these studies. Possible processes which account for the observed changes and the relationship of these changes to combustion and ballistic properties are discussed. The compressive strengths of all unaged candidate LOVA propellants investigated are significantly lower than the compressive strengths of unaged NC single, double, and triple base propellants at  $-45^{\circ}\mathrm{C}$  and most NC propellants at  $23^{\circ}\mathrm{C}$ .

# ACKNOWLEDGMENT

The authors are indebted to members of the Gun Propulsion Technology Branch, Applied Science Division for organization and coordination of the program of which the results reported here are a part. Particular thanks are due to B. Strauss and P. Hui for many helpful discussions and for making samples available. Thanks are also due to T. Richter for several helpful discussions. Finally, M. Mezger is thanked for experimental assistance and contributions to data analysis.

# CONTENTS

		Page
Introduction		1
Experimental	,	1
Propellants Studied Sources of Scatter and Errors in Results		2 2
Results		3
General Candidate LOVA Propellants Nitrocellulose Base Propellants		3 3 5
Conclusions		9
References		11
Distribution List		21

# TABLES

		Page
1	LOVA candidate propellant compositions	13
2	Standard NC propellant compositions	14
3	Propellant testing schedule and test temperatures (°C)	15
4	Compressive strengths (psi) measured at $-45^{\circ}\text{C}$ of candidate LOVA propellants treated at $65.5^{\circ}\text{C}$ for various time periods	16
5	Comrpessive strengths (psi) measured at 23°C of candidate LOVA propellants treated at 65.5°C for 0 and 18 months	17
6	Compressive strengths (psi) measured at -45°C of standard nitrocellulose base propellants treated at 65.5°C for various time periods	18
7	Compressive strengths (psi) measured at 23°C of standard nitrocellulose base propellants treated at 65.5°C for 0 and 18 months	19
8	Compressive strengths (psi) measured at 23°C of M-30 propellant treated at 65.5°C for various time periods	20

#### INTRODUCTION

The purpose of this overall program is to develop low vulnerability (LOVA) propellants, and the objective of this particular program has been to determine the effect of high temperature accelerated aging on several properties of candidate LOVA propellants and to compare these effects with those of standard nitrocellulose (NC) base propellants subjected to the same treatment. Thermal treatment at 65.5°C was chosen at least in part because this temperature is in use for safety tests of some NC base propellants. Only the effects of thermal treatment on mechanical properties are discussed here. The effects of this thermal treatment on other properties are discussed elsewhere (refs 1, 2, and 3).

During ignition and combustion propellant grains are subjected to extremely high pressures for very short times. All of the materials are visco-elastic, i.e., their properties are rate and temperature dependent, and may become extremely brittle at low temperatures and/or high loading rate. When ignited, the high pressures and pressure gradients produced in a gun chamber can cause mechanical breakup of the propellant grains which would lead to erratic burning. Many of these propellants are two- or three-phase mixtures and contain binder and plasticizer, one or both of which may be energetic. They are stored over long periods of time at a wide range of temperatures. Over long periods of time and under extreme temperature conditions, changes, such as decomposition of the binder, diffusion, decomposition of the plasticizer or changes in the binder/plasticizer interaction might occur which effect mechanical properties in particular, and physical properties in general. Therefore, this accelerated aging program was undertaken to monitor possible changes in these properties under these con-Measurements were made after several time periods at 65.5°C up to 18 ditions. months at this temperature.

#### EXPERIMENTAL

Experimental information has been presented (ref 4) and a detailed description of the equipment and experimental techniques will be given elsewhere (ref 5). A few pertinent experimental details not given previously (ref 4) are given in this section. The computer controlled servo-hydraulic loading system was operated in stroke control and programmed for 50% maximum engineering strain Because of the response time of the with a square wave displacement input. system and sample, this resulted in times to failure, or maximum compressive stress of 2 to 10 ms, and times to 50% strain of approximately 25 ms. In all cases complete load and displacement versus time data were taken, printed, and Engineering and true stresses and strains were curves plotted via computer. calculated and stress-strain curves plotted. Only engineering stresses are reported here because conditions were such that there is negligible difference between engineering and true stresses. Strains are not reported because of possible calibration problems which have yet to be resolved; however, comments are given on the general nature of the stress-strain curves. Comments on sample after measurement are for samples subjected to 50% compression; condition

however, in most cases at low temperature fracture and fragmentation occured at much lower strains. In general measurements were made in sequence, i.e., one sample of each propellant was measured for a given set of conditions before a second sample of each propellant was measured. This was done as part of an attempt to eliminate systematic errors. Because of the large number of measurements required for this program and limited time and resources available it was necessary to compromise on types and in some cases precision of data. For example, sample fragments were not collected for each aging period and the scatter in the results could have been reduced by allotting more time for each sample measurement.

# Propellants Studied

The LOVA propellants and their compositions are given in table 1, and five of the seven NC propellants and their compositions are given in table 2 (ref 4). The two NC propellants not listed, F527 and NQ, are both United Kindgom propellants. They are both triple base and similar in composition to M30. The one significant difference between M30 and F527 is that F527 has cyclotrimethylenetrinitramine (RDX) in place of approximately 50% of the NQ. Measurements were made after 0, 1, 2, 3, 6, 12, and 18 months storage at 65.5°C. Not all propellants were measured after each aging period. A schedule of propellants measurements were made at -45°C, but limited measurements were made at 23°C as indicated in table 3. All propellants except HYCAR with and without stabilizer, NQ, and F527 were measured after 18 months at both temperatures. Samples of these four propellants were not available for measurement at this time.

In a separate study, designated Study II, cellulose acetate butyrate/nitrocellulose/acetyl triethyl citrate (CAB/NC/ATEC), M-30, and CAB/ATEC were subjected to the same aging conditions and measurements were made only at -45°C after 0, 3, 6, and 12 months. The results for CAB/ATEC are included in table 4. The results for CAB/NC/ATEC for Study II are discussed below. The CAB/NC/ATEC used for the two studies was produced at different locations. The composition of CAB/ATEC is 16% CAB, 8% ATEC, 75% RDX, and 1% potassium sulfate (K $_2$ SO $_4$ ). All of the above propellant samples were in the form of multiperforated grains. Measurements were also made on unaged rods of CAB/NC/ATEC and cellulose acetate proprionate/nitrocellulose/acetyl triethyl citrate (CAP/NC/ATEC).

#### Sources of Scatter and Errors in Results

As discussed in reference 4, poor temperature control and inadequate temperature measurement during low temperature conditioning and measurement in the early stages of the program were thought to be primary causes of the considerable scatter in the results. Additional measurements and analysis suggest that this is not the case. Unaged (0 month) samples of all NC base and most LOVA propel-

lants were measured after considerable improvements were made in temperature control and measurement without significant changes in the results. In addition, the scatter in the data was not significantly reduced for aged propellants measured after these changes were introduced. Samples of unaged M-30 were also measured as a function of temperature. The variation of compressive strength with temperature is not sufficient to account for the observed scatter in the data for the extremes of temperature that could reasonably be expected to have occured. The temperature dependence of the compressive strengths for the other propellants between 23°C and -45°C are not known. However, the propellants with the greatest percentage change between the two temperatures do not necessarily show the greatest scatter in results. All of the above strongly indicates that poor temperature control and measurement is not the primary cause of uncertainty in the results.

As noted in reference 4, another cause of scatter in the results is stress concentrations due to nonparallel and nonflat ends. In recent work with other propellant type materials, good reproducibility was obtained by carefully working the sample ends after machining to ensure flat parallel surfaces (ref 6). Strain induced by temperature gradients and thermal cycling may also contribute to lack of reproducibility. However, studies to date indicate that the major causes of scatter are poor sample end conditions as discussed above and inherent properties of the propellant grains. The latter may include nonhomogeneous sample composition, variations in sample (grain) perforations, and grain curvature. Variations in temperature may contribute in part to lack of reproducibility. Extreme values of compressive strength, mostly low values, are not included in the results presented.

#### RESULTS AND DISCUSSION

General

Compressive strengths at  $-45^{\circ}\text{C}$  are reported after 0, 1, 6, 12, and 18 months aging at 65.5°C. While limited measurements were made at 1, 2, and 3 months aging, the results indicate little or no change for these aging times (see below). Although measurements were made at  $-45^{\circ}\text{C}$  and  $23^{\circ}\text{C}$ , most measurements were made at the lower temperature as discussed in reference 4. Compressive strengths at  $23^{\circ}\text{C}$  after 0 and 18 months aging are also reported. Again, limited measurements were made at  $23^{\circ}\text{C}$  after 1, 2, and 3 months and the results indicate little or no change (see below).

# Candidate LOVA Propellants

In table 4, compressive strengths, standard deviations, and numbers of samples measured at -45°C are given as a function of aging time at 65.5°C. Although there are some discrepancies and unexplained observations, the results do not

indicate significant trends for changes in compressive strength for most propellants with aging within the standard deviations which are large. the two HYCAR's, and CA/TA compressive strengths are unchanged after 18 months aging. The same applies to CAP/NC/ATEC except for an anomalously low value at 12 months. Examination of the stress-strain curves does not give insight into this low value and it is tentatively concluded that poor samples were measured for this aging time. While the compressive strength of CAB/TA is unchanged between 6 and 18 months, there is insufficient data to make any conclusions about changes from 0 to 6 months. The data for CAB/ATEC (Study II) indicates a decrease at 12 the data for ethyl cellulose/nitrocellulose/dibutylphthalate (EC/NC/DBP) suggests a decrease over the 18 month period although the standard deviations are large. Additional measurements are necessary in particular for CAP/NC/ATEC and CAB/TA, and are also desirable for EC/NC/DBP to clarify, complete, and substantiate the results to date.

In a separate study (Study II) the compressive strength of CAB/NC/ATEC did not change over a 12 month aging period. However, the compressive strength of this propellant which was made at a different location is approximately 1.8 times the values given in table 4. Most of the propellants of table 4 were made at the same location as the CAB/NC/ATEC of Study II. Exceptions are CAP/NC/ATEC and EC/NC/DBP. A difference in RDX particle size distribution (ref 7) and processing procedures may account for the difference between the two CAB/NC/ATEC's made at different locations.

In table 5 results are given for measurements at  $23^{\circ}\text{C}$  for 0 and 18 months aging times. The CAP/NC/ATEC, CAB/NC/ATEC, and EC/NC/DBP propellants have the same compressive strengths at 0 and 18 months. The data suggests that the strength of CAB/TA increases somewhat and the strength of CA/TA decreases somewhat with 18 months aging. These comments regarding changes must be qualified by the fact that only 3 samples of each propellant were measured at each aging time. Samples of the two HYCAR propellants were not available after 18 months aging.

Additional information is obtained from the stress-strain curves and the sample condition after compression. All of the candidate LOVA propellants failed by fracture at  $-45\,^{\circ}\text{C}$ . The decrease of the stress after the maximum stress had been attained is more abrupt as a function of stress at 18 months compared to 0 months. In addition, these samples tended to fail by cracking, fracture, and fragmentation at zero and short aging times, but to completely fragment into small pieces after 18 months aging. Thus changes in the  $-45\,^{\circ}\text{C}$  stress-strain curves and in the sample conditions indicate increased brittleness even in cases where the compressive strength is not changed appreciably.

At -45°C the stress, as a function of strain increases to the maximum stress, (at approximately 2% to 10% strain) and then decreases rather sharply and approaches zero. In contrast at 23°C, the stress as a function of strain increases to the maximum stress and then decreases more or less gradually up to the 50% programmed compression. While observations have not been made during this rapid compression, observations of sample conditions after compression indicate that barrelling, cracking, and fragmentation take place during compression at 23°C. Barrelling occurs because of friction between the sample and support

and cracking apparently occurs at least in part because of this barrelling. A more complete description of the failure processes will be given elsewhere (ref 8).

At 23°C the stress values after the maximum stress are in general larger at 0 months than at 18 months for a given strain thus indicating more failure after 18 months aging. The CAP/NC/ATEC and CAB/NC/ATEC propellants are exceptions to this and show the opposite effect with aging. All LOVA propellants were found to have cracked and broken into large pieces after 50% compression. The degree of fracture was greater and the size of the pieces was smaller at 18 months as compared to 0 months. Thus at 23°C changes in the stress-strain curves and failure were observed even in cases where the compressive strength did not change appreciably.

In summary the compressive strengths of most candidate LOVA propellants investigated at -45 and 23°C do not change as a result of aging at 18 months at 65.5°C. The standard deviations are large and in cases where changes may have occurred, e.g., EC/NC/DBP at  $-45^{\circ}$ C, additional measurements are desirable to support the results to date. In other cases, e.g., at 23°C, more measurements are required to confirm the results presented. While the compressive strengths are not significantly altered with aging the failure mode is altered as evidenced by changes in the stress-strain curves and the degree of fracture and fragmenta-In general there is a tendency toward increased brittleness with aging for Increased brittleness could be due to loss of plasticizer and/or changes in the binder or binder-plasticizer interaction. Clearly additional measurements of such parameters as plasticizer content, binder molecular weight, and degree of cross-linking are necessary to understand these changes. Mechanical properties studies as a function of temperature and studies of the glass transition temperature  $(T_G)$  will also aid in understanding these results. Changes in EC/NC/DBP may be due to decomposition of NC since there is stabilizer depletion (refs 3 and 7). However, the results of studies of NC base propellants (below) suggest that this is not the case.

# Nitrocellulose Base Propellants

In table 6 the compressive strengths, standard deviations and number of samples measured are given at -45°C for 0, 1, 6, 12, and 18 months aging at 65.5°C. Measurements were made on single, double and triple base propellants. Compositions are given in reference 4. The results in table 6 show that the compressive strengths of the two single base propellants, NACO, and M6+2 do not change with this aging. In contrast the compressive strength of the standard double base propellant M26 decreases by approximately a factor of three with 18 months aging. The results for another double base propellant, JA2, are not as clear. There is a significant decrease of the compressive strength at 12 months as previously reported (ref 4), but an increase at 18 months to a value similar to the original value. The JA2 propellant contains two plasticizers, nitroglycerin (NG), and diethylene glycol dinitrate (DEGDN) whereas M26 contains only NG. Additional results and discussion of JA2 are given below. Three triple base

propellants, M-30, NQ, and F527, were also investigated. The compressive strength for M-30 decreases by approximately a factor of two after 18 months aging. Samples of NQ and F527 were not available for measurement at 18 months. However, the trend of a decrease of compressive strength with aging time is clear.

In table 7 similar results are given for measurements at 23°C for 0 and 18 months for most NC propellants except M-30. The results for M-30 are given in table 8 for 0, 1, 2, 3, and 18 months. As with the candidate LOVA propellants the significance of the 23°C results are limited because only three measurements were made for each case. Within these limitations the compresive strength of the single base propellants NACO and M6+2 and the double base propellant M26 are not changed within limits of 5 to 10%. The "modified" double base propellant JA2 gave the same compressive strength at 0 and 18 months. Of the triple base propellants, only M-30 was measured at several aging periods other than 0 months. The results for M-30 as given in table 8 suggest but do not establish a gradual decrease of the compressive strength with increasing aging time. However, all of these apparent changes must be confirmed by additional measurements.

As with the candidate LOVA propellants observation of the general nature of the stress-strain curves and sample conditions after compression give additional information regarding the effects of aging on the NC base propellants. unaged single base propellants NACO and M6+2, at -45°C the stress as a function of strain increases to the maximum stress, and then gradually decreases with increasing strain and finally moderately abrupt failure is observed at large In contrast, after 18 months aging an abrupt decrease in stress was The samples after compression were found to have found after maximum stress. changed from barrelling and cracking with some fracture at 0 months to fracture into large pieces at 18 months. Thus changes in the stress-strain behavior and sample conditions after compression were found although no change in the compressive strength was detected as is the case for some LOVA propellants. The stressstrain curves for the double base propellant M26 show a moderately abrupt decrease of the stress after the maximum stress was reached for 0 months but a more abrupt decrease of stress after maximum stress at 18 months. fractured into large pieces at both aging times. The double base propellant JA2 stress-strain curves at 0 months are similar to those of the single base propel-While the stress-strain curves at 6 and 12 months show essentially the same characteristics as the 0 month curves, the curves at 18 months indicate rather abrupt failure at the compressive strength. These results suggest that the decrease of the compressive strength at 12 months (table 6) is due to the same processes which cause the decrease for the other NC base propellants (see below) but that the increase at 18 months is due to either a continuation of this process and not observed for other propellants or to additional processes. Clearly additional work is required to substantiate the results and for under-After all ages after compression at -45°C cracking and some fracture standing. were found. The stress-strain curves for the triple base propellants M-30, F527, and NQ show essentially the same characteristics as the double base propellant M26 at 0 months and the same changes with aging, i.e., a more abrupt decrease of the stress at failure. Observations of M-30 after compression indicate somewhat more complete fragmentation at 18 months compared to 0 months. Although samples of NQ and F527 were not measured at 18 months, all three triple base propellants exhibited considerable fracture at 0 months.

At 23°C no significant changes from 0 to 18 months were observed in either the stress-strain curves or the samples after compression for the single base propellants NACO and M6+2. Stress as a function of strain increased to the maximum and then decreased slightly with increasing strain. Maximum compression (50%) caused barrelling and axial cracking. Some changes were observed for the double base propellant M26. The stress-strain curves indicate a lower stress for a given strain at 18 months compared to 0 months. Barrelling and cracking were found at 0 months but barrelling, cracking, and some fracture were observed at 18 In contrast, no significant changes were observed in the stress-strain curves or sample condition after compression for the double base propellant JA2. The stress-strain curves are somewhat similar to those for the single base propellants, but barrelling and little or no cracking were found after compression. The results for M-30 are also similar to those for the single base propellant, i.e., no significant change in the stress-strain curves and barrelling and cracking as a result of compression at both 0 and 18 months.

In summary, only M26 at 23°C shows changes in the stress-strain curves and sample conditions when comparing 0 and 18 months aging. Within the precision of the results the compressive strengths of the NC propellant are also unchanged with the possible exception of M-30 as noted above. At -45°C all NC base propellants show some changes as a result of aging. The compressive strengths of the double and triple base propellants are decreased and the stress-strain curves and sample conditions after compression indicate increased brittleness for all NC propellants investigated. In order to understand these changes it is desirable to have some insight into the parameters which determine the mechanical properties before aging. A discussion of these factors is given before a discussion of possible causes of the changes.

All NC base propellants with the exception of JA2 have approximately the same compressive strength at -45°C, i.e., 30,000 to 35,000 psi as given in table 6. JA2 is made by a solventless process whereas all of the other propellants in the group are made by a solvent process. This difference in processing may be at least a partial cause of the lower compressive strength of JA2. Measurements on experimental samples containing approximately 95% NC and 5% total volatiles indicate compressive strengths in the range of the samples in table 6 at  $-45\,^{\circ}\text{C}$ (except JA2) and in addition values at 23°C which are in agreement with the values for the single base propellants NACO and M6+2 (ref 8). These results suggest that the compressive strengths of the single base propellants at 23°C and the single, double, and triple base propellants of table 6 at -45°C are primarily The NC samples referred to above were made from NC of two determined by the NC. different percent nitrations, and different solvents were used in the processing (ref 9). The double base propellant M26 and the triple-base propellants M-30 and NO have a lower and similar compressive strength at 23°C. Experimental samples of NC/NG containing 85% NC and 15% NG gave compressive strengths at -45 °C and 23°C which are very close to the values obtained for some double and triple base These propellants and the experimental NC/NG samples have propellants (ref 8). It is thus concluded that the lower compressive in common the plasticizer NG. strength at 23°C in NG containing materials is due to the plasticizing action of NG. The JA2 propellant, which contains NG, has a second plasticizer and has an even lower compressive strength at 23°C (see table 7). Although F527 contains

only NG as a plasticizer it also has a lower compressive strength at 23°C. lower compressive strength may be due to the presence of RDX. Another triple base propellant containing RDX was previously found to give a lower compressive strength at room temperature (refs 10, 11, and 12). It is thus tentatively concluded that the compressive strengths of NG containing NC propellants at 23°C is determined by NC as modified by the plasticizing action of NG and that at -45°C this plasticizing action is "frozen out" so that the compressive strength is determined primarily by the NC. The strength of non-NG containing material is determined by the NC at both -45 and 23°C. The JA2 and F527 propellants have different compressive strengths because of differences in composition and This discussion is applicable only for the strain rates used in processing. these experiments. The results further suggest that the fracturing of triple and double base propellants (except JA2) at -45°C after the maximum stress has been reached is due to the presence of NG since unaged single base propellants generally do not fracture at this temperature. It is rather surprising that the triple base propellants have properties similar to the double base M26, thus indicating that the rather high filler content of the triple base propellants does not significantly influence the properties measured. The addition of a second plasticizer and/or differences in processing appears to have a desirable effect since JA2 was not observed to fracture for the conditions of these experiments.\*

Based on this discussion regarding possible rationale for the mechanical properties of unaged NC base propellants it is possible to address reasons for the observed changes upon aging (heat treatment). Both NC and NG are known to thermally decompose and heat treatment at 65.5°C is part of a standard safety test for single and double base propellants (refs 14). The decompositon of NC is autocatalytic and all NC base propellants contain a stabilizer to hinder this autocatalytic action. The stabilizer concentration of the NC propellants was measured as a function of aging time and very significant stabilizer depletion was found after 18 months indicating decomposition (refs 1 and 7). It is also known that the products of NG decomposition can cause NC decomposition (ref 14). This appears to be a significant factor because the compressive strength of only NG containing propellants was decreased by aging, i.e., M26, M30, NQ, F527, and Because there is no significant change in the compressive apparently JA2. strength of the two single base propellants NACO and M6+2 as a result of thermal treatment (aging), it is concluded that NC does not undergo major changes when no Since there is significant stabilizer depletion in these NG is present. propellants there must be some NC decomposition and this may be the cause of the increased embrittlement at 18 months. Thus, the decrease of the compressive strength at -45°C for the double and triple base propellants is tentatively attributed to NC decomposition caused by NG decomposition and or NG diffusion or The lack of significant change in properties at 23°C suggests that there is sufficient NG remaining to produce the plasticizing effect and that this

<sup>\*</sup>JA2 has been found to fracture at lower temperatures and/or higher strain rates (refs 5 and 13).

dominates the measured mechanical properties at this temperature. Measurements at intermediate temperatures for both unaged and aged materials are highly desirable and would aid in understanding of the observed phenomena. Measurement of the glass transition temperature, plasticizer content, NC molecular weight distribution, etc. should further aid in understanding. The decrease of the compressive strength for JA2 at 12 months may be due to the above mentioned effects of NG decomposition on NC but the increase at 18 months may be due to more significant changes in composition.

In summary the compressive strengths of unaged NC base propellants at -45°C are apparently primarily due to the properties of NC and the changes which have been observed at this temperature as a result of heat treatment are postulated to be due to NC decomposition caused by the products of NG decomposition or NG diffusion. The compressive strengths at 23°C are determined by NC in propellant not containing NG and by the plasticizing effect of NG on NC in propellants containing NG. Changes in the compressive strength at this temperature were not observed because there has not been sufficient NG/NC decomposition to significantly alter this plasticizing effect. The embritlement observed at -45°C in all NC base propellants may also be due to NC decomposition. It is not possible to explain all of the observations with the information available. This interpretation is highly speculative and additional experimentation is necessary to provide confirmation.

#### CONCLUSIONS

For the conditions of measurements used in this work the mechanical properties of candidate LOVA propellants are much more stable with accelerated aging at 65.5°C than double and triple base NC propellants. Changes in single base NC propellants are similar to those observed in the LOVA propellants for this heat treatment. Large decreases in the compressive strength of double and triple base NC propellants have been observed. While the results to date suggest that these changes are associated with NG decomposition or diffusion, additional experiments are required to establish the causes of the changes found. The compressive strengths of all unaged candidate LOVA propellants investigated are significantly lower than the compressive strengths of unaged NC single, double, and triple base propellants at -45°C and most NC propellants at 23°C. In addition, the LOVA propellants tend to exhibit greater brittleness than NC base propellants.

The decreased compressive strength of aged double and triple base propellants at -45°C means that grain fracture under gun firing conditions can occur under less severe mechanical loading and thus represents a greater hazard in terms of propellant charge malfunctions in which mechanical failure plays a role. Charge malfunctions can lead to erratic ballistics and breechblows. The same considerations apply to the lower compressive strengths of unaged candidate LOVA propellants and the tendency for greater brittleness compared to unaged NC base propellants.

#### REFERENCES

- 1. B. Strauss, P. Hui, S. Wise, A. Beardell, E. Costa, L. Avrami, C. Jamison, and J. Pinto, "Aging Study of Low Vulnerability (LOVA) Propellants," ARLCD-TR-83010, April 1983.
- 2. B. Strauss, P. Hui, A. Beardell, and E. Costa, "Burning Rate Performance of Insensitive Explosives as a Function of Aging," 1982 JANNAF Combustion Meeting, CPIA Publication 366, October 1982.
- 3. M Kirshenbaum, L. Avrami, and B. Strauss, "Sensitivity Characterization of Low Vulnerability (LOVA) Propellants," ARLCD-TR-83005, March 1982.
- 4. J. Pinto, S. Nicolaides, D. Georgevich, and D.A. Wiegand, "Mechanical Properties of Candidate LOVA and Other Gun Propellants as a Function of High Temperature Accelerated Aging," CPIA Publication 366, October 1982.
- 5. J. Pinto, S. Nicolaides, and D.A. Wiegand, to be published.
- 6. M. Mezger, J. Pinto, and D.A. Wiegand, unpublished results.
- 7. B. Strauss, private communication.
- 8. M. Mezger, J. Pinto, and D.A. Wiegand, to be published.
- 9. B. Lehman, private communication.
- 10. S. Nicolaides, D.A. Wiegand, and J. Pinto, "The Mechanical Behavior of Gun Propellant Grains and Its Role in Interior Ballistics," 16th JANNAF Structure and Mechanical Behavior Meeting, CPIA Publ. 311, Vol. I, December 1979.
- ll. S. Nicolaides, J. Pinto, and D.A. Wiegand, "Mechanical Properties, Mechanical Grain Failure, and Changes in the Burning Performance of Gun Propellants," JANNAF Propulsion Meeting, CPIA Publ, 315, Vol. I, March 1980.
- 12. S. Nicolaides, D.A. Wiegand, and J. Pinto, "The Mechanical Behavior of Gun Propellant Grains in Interior Ballistics," ARLCD-TR-82010, June 1982.
- 13. R.J. Lieb and J.J. Roccio, "High Strain Rate Mechanical Properties Testing on Lots of Solid Gun Propellant with Deviant Interior Ballistic Performance," CPIA Publ. 368, 23 (1982).
- 14. T. Richter, private communication.

Table 1. LOVA candidate propellant compositions

			CAB/NC/	CAP/NC/	EC/NC/		
Component	CA/TA	CAB/TA	ATEC	ATEC	DBP	HYCAR	HYCAR/STAB
RDX	75.0	75.0	76.0	<b>76.</b> 0	73.5	80.0	80.0
CA	16.0	,					
CAB		16.0	12.0				
CAP				12.0			
EC					11.7		
NC (% nitration)			4.0 (12.6)	4.0 (12.6)	7.4 (12.6)		
HYCAR						18.3	18.3
Triacetin	8.0	8.0					
ATEC			7.6	7.6			
DBP					6.7		
Ethyl centralite			0.4	0.4	0.7		
K <sub>2</sub> SO <sub>4</sub>	1.0	1.0					
TP 95						1.0	1.0
Graphite						0.7	0.7
Staboxol							0.15
Weston 618 antiozonate							0.15
Tollanox					0.515		

Table 2. Standard NC propellant compositions

	<u>M30</u>	<u>M26</u>	<u>M6+2</u>	NACO	JA2
NC (% nitration)	27.61 (12.61)	66.10 (13.15)	86.77 (13.15)	93.61 (12.0)	63.5 (13.0)
NG	22.67	25.80			14.0
NQ	47.96				
Ethyl centralite	1.49	6.35		1.15	
Cryolite	0.27				
Graphite	0.17*	0.36			0.05
Barium nitrate		0.71			
Potassium nitrate		0.68			
Dinitrotoluene		s vm da	9.60		
Diphenylamine			1.10**		
Potassium sulfate			2.09**	1.20	
Lead carbonate		100 mil 100		1.14	
Butyl stearate				2.90	
Total volatiles	0.50		um vid rui	2.63	
DEGDN		-100 VIII			21.7
Akardit II					0.7
Magnesium oxide					0.05
DBP			3.61		

<sup>\*</sup>Added as glaze

<sup>\*\*</sup>Added

Table 3. Propellant testing schedule and test temperatures (°C)

			Te	mperature	(°C)		
Propellant	Unaged	1 mo.	2 mo.	3 mo.	6 mo.	12 mo.	18 mo.
CA/TA	23/-45	23/-45	-	-	-45	<del>-</del> 45	23/45
CAB/TA	23/-45	23/-45	23/-45	-	-45	<b>-</b> 45	23/-45
EC/NC/DBP	23/-45	23/-45	23/-45	23/-45	<del>-</del> 45	-45	23/-45
CAB/NC/ATEC	23/-45	23/-45	-	-45	<b>-</b> 45	<b>-</b> 45	23/-45
CAP/NC/ATEC	23/-45	23/-45		-45	<b>-</b> 45	<b>-</b> 45	23/-45
HYCAR	23/-45	23/-45	-	-45	-45	-45	-
HYCAR/STAB	23/-45	23/-45	-	<b>-4</b> 5	<b>-</b> 45	-45	-
M-30	23/-45	23/-45	23/-45	23/-45	-45	<b>-</b> 45	23/-45
NACO	23/-45	23/-45	-	-	-45	-45	23/-45
NQ	23/-45	23/-45	-	-	-45	-45	
M6+2	23/-45	23/-45	-	- ,	-45	-45	23/-45
JA2	23/-45	23/-45	-	-	-45	-45	23/-45
F527	23/-45	23/-45	-		-45	-45	-
M26	23/-45	23/-45	-		-45	-45	23/-45

Compressive strengths (psi) measured at  $-45^{\circ}$ C of candidate LOVA propellants treated at 65.5°C for various time periods Table 4.

	CA/TA	CAB/TA	CAB/ NC/ATEC	CAP/ NC/ATEC	EC/NC/DBP	HYCAR/ STAB	HYCAR	CAB/ATEC
0 + 1 MONTH AVERAGED <sup>a</sup>								
Compressive Strength Std Dev <sup>b</sup> Number <sup>C</sup>	14,200 4,800 7	19,800 1,900 2	12,300 2,800 6	15,000 2,600 7	16,000 2,600 9	13,200 12,800 2,300 2,600 8 7	12,800 2,600 7	16,300 1,500 5
6 MONTHS								
Compressive Strength Std Dev Number	13,500 4,000 7	13,600 3,600 6	10,400 1,300 5	15,000 1,400 5	14,600 5,000 6	11,500 12,600 1,200 2,800 5 5	12,600 2,800 5	18,600 3,000 5
12 MONTHS								
Compressive Strength Std Dev Number	17,800 2,700 6	14,500 6,300 7	11,300 1,700 7	8,800 1,100 5	13,400 3,400 7	11,100 12,700 2,900 1,400 5 5	12,700 1,400 5	11,300 2,300 6
18 MONTHS								
Compressive Strength Std Dev Number	15,200 2,400 6	14,500 2,600 4	13,600 2,500 7	14,200 1,700 5	9,300 1,200 5			

<sup>&</sup>lt;sup>a</sup>Since 0 and 1 month data showed no difference, averages of the two are shown here for all propellants except CAB/ATEC (Study II) for which I month measurements were not made.

bstandard Deviation calculated according to the formula  $\left[\frac{1}{N-1}\right]_{1}^{N}\left[\frac{1}{\sigma}-\sigma\right]^{2}^{1/2}$ 

<sup>&</sup>lt;sup>C</sup>Number of samples measured

Compressive strengths (psi) measured at  $23^{\circ}\mathrm{C}$  of candidate LOVA propellants treated at  $65.5^{\circ}\mathrm{C}$  for 0 and 18 months Table 5.

	CA/TA	CAB/TA	CAB/NC/ATEC	CAP/NC/ATEC	EC/NC/DBP	HYCAR/STAB	HYCAR
O MONTH							
Compressive Strength	13,900	9,200	7,300	6,500	8,500	2,600	2,800
Std Dev <sup>a</sup>	2,800	2,500	1,500	1,500	300	300	9
Number	n	า	n	n	n	n	)
18 MONTHS							
Compressive Strength	10,500	13,900	7,400	8,500	7,100		
Std Dev	200	006	1,500	3,400	1,800		
Number	က	က	e	က	က		

<sup>a</sup>Standard Deviation <sup>b</sup>Number of samples measured

Compressive strengths (psi) measured at -45°C of standard nitrocellulose base propellants treated at 65.5°C for various time periods Table 6.

F527 M26 JA2	29,800 34,600 23,000 3,400 6,000 3,600 7 5	21,400 21,800		23,200 22,400 21,200 5,600 6,400 1,500 7 7	23,600 11,100 13,600 1,600 3,100 2,100 5 6	10,100 22,700 2,600 2,800 5
NQ	31,000 2,500 6	31,500	) '\	27,500 2,500 5	23,600 4,200 7	
M30	34,000 4,600 6	30,600	7	24,900 6,800 8	25,700 3,300 9	15,800 6,400 8
	Compressive Strength Std Dev <sup>a</sup> Number <sup>b</sup>	MONTH   Compressive Strength		Compressive Strength Std Dev Number	Compressive Strengths Std Dev Number	Strength

 $^{\mathbf{a}}_{\text{Standard Deviation}}^{\mathbf{a}}_{\text{Number of samples measured}}$ 

Compressive strengths (psi) measured at  $23^{\circ}\text{C}$  of standard nitrocellulose base propellants treated at  $65.5^{\circ}\text{C}$  for 0 and 18 months Table 7.

	O MONTH	Compressive Strength 11 Std Dev <sup>a</sup> Number <sup>b</sup>	18 MONTHS	Compressive Strength Std Dev Number
NQ		11,300 500 3		
F527		4,000 800 3		
M26		8,900 1,400 2		10,100 600 3
JA2		2,400 500 3		2,400 200 3
NACO		16,900 200 3		19,500 2,100 3
M6+2		18,900 600 3		22,000 1,000 3

<sup>a</sup>Standard Deviation <sup>b</sup>Number of samples measured

Compressive strengths (psi) measured at  $23^{\circ}\text{C}$  of M-30 propellant treated at  $65.5^{\circ}\text{C}$  for various time periods Table 8.

	0 Month	1 Month	2 Months	3 Months	18 Months
Compressive Strength	11,900	10,500	006,6	10,100	8,500
	700	200	800	300	006
		3	e	3	33

 $^{\rm a}{\rm Standard}$  Deviation  $^{\rm b}{\rm Number}$  of Samples Measured

### DISTRIBUTION LIST

Commander

Armament Research and Development Center

U.S. Army Armament, Munitions and Chemical Command

ATTN: DRSMC-GCL(D)

DRSMC-LCA-G(D), B. Strauss

DRSMC-LCE(D) (3)
DRSMC-LCE-C(D), 20
DRSMC-TSS(D) (5)

Dover, NJ 07801

Administrator

Defense Technical Information Center

ATTN: Accessions Division (12)

Cameron Station

Alexandria, VA 22314

Director

US Army Materiel Systems Analysis Activity

ATTN: DRXSY-MP

Aberdeen Proving Ground, MD 21005

Commander/Director

Chemical Research and Development Center

U.S. Army Armament, Munitions and Chemical Command

ATTN: DRSMC-CLJ-L(A)
DRSMC-CLB-PA(A)

APG, Edgewood Area, MD 21010

Commander

US Army Armament, Munitions and Chemical

Command

ATTN: DRSMC-LEP-L(R)

Rock Island, IL 61299

Director

Ballistics Research Laboratory

Armament Research and Development Center

U.S. Army Armament, Munitions and

Chemical Command

ATTN: DRSMC-BLA-S(A)

DRSMC-BLI(A), R. Lieb

DRSMC-LCB-TL(A)

Aberdeen Proving Ground, MD 21005

Director

U.S. Army TRADOC Systems Analysis Activity

ATTN: ATAA-SL

White Sands Missile Range, NJ 88002

Chief
Benet Weapons Laboratory, LCWSL
Armament Research and Development Center
U.S. Army Armament, Munitions and Chemical Command
ATTN: DRSMC-LCB-TL
Watervliet, NY 12189

Lawrence Livermore National Laboratory ATTN: M.S. Castantino P.O. Box 808 Livermore, CA 94550